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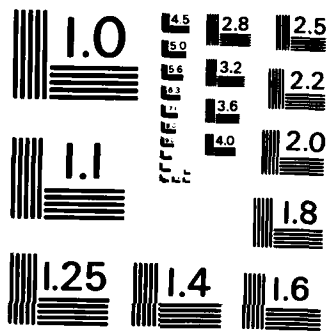

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August 30, 1985

Final Scientific Report

PRODUCTION AND PROPERTIES OF METASTABLE AUTODETACHING NEGATIVE IONS

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Prepared for:

U.S. AIR FORCE OFFICE OF SCIENTIFIC RESEARCH
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SRI Project No. PYU-4485
Contract No. F49620-82-K-0030
MP 85-203

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Unclassified

SECURITY CLASSIFICATION OF THIS PAGE

REPORT DOCUMENTATION PAGE

1a. REPORT SECURITY CLASSIFICATION Unclassified	1b. RESTRICTIVE MARKINGS None
2a. SECURITY CLASSIFICATION AUTHORITY N/A	3. DISTRIBUTION/AVAILABILITY OF REPORT Unrestricted
2b. DECLASSIFICATION/DOWNGRADING SCHEDULE N/A	
4. PERFORMING ORGANIZATION REPORT NUMBER(S) MP 85-203	5. MONITORING ORGANIZATION REPORT NUMBER(S) AF
6a. NAME OF PERFORMING ORGANIZATION SRI International	6b. OFFICE SYMBOL (If applicable)
7a. NAME OF MONITORING ORGANIZATION Air Force Office of Scientific Research	
6c. ADDRESS (City, State and ZIP Code) 333 Ravenswood Ave. Menlo Park, CA 94025	7b. ADDRESS (City, State and ZIP Code) Bolling Air Force Base Washington, DC 20332
8a. NAME OF FUNDING/SPONSORING ORGANIZATION AFOSR	8b. OFFICE SYMBOL (If applicable) NP
9. PROCUREMENT INSTRUMENT IDENTIFICATION NUMBER F49620-82-K-0030	
8c. ADDRESS (City, State and ZIP Code) Bolling AFB D.C. 20332	10. SOURCE OF FUNDING NOS.

PROGRAM ELEMENT NO.	PROJECT NO.	TASK NO.	WORK UNIT NO.
61102F	2301	A7	

11. TITLE (Include Security Classification)
Production & Properties of Metastable Autodetaching Negative Ions

12. PERSONAL AUTHOR(S)
James R. Peterson and Michael J. Coggiola

13a. TYPE OF REPORT	13b. TIME COVERED	14. DATE OF REPORT (Yr., Mo., Day)	15. PAGE COUNT
Final Scientific	FROM 82/5/15 TO 85/6/14	1985 Aug. 20	22

16. SUPPLEMENTARY NOTATION
The findings in this report are not to be construed as an official Department of the Air Force position, unless so designated by other authorized documents.

17. COSATI CODES			18. SUBJECT TERMS (Continue on reverse if necessary and identify by block number)
FIELD	GROUP	SUB. GR.	Negative ions, beams, metastable, autodetachment, photodetachment, resonances.

19. ABSTRACT (Continue on reverse if necessary and identify by block number)

Experimental work has been directed primarily toward determining properties of metastable autodetaching negative ions, with a smaller effort directed to their production in beams, as well as the production and photodetachment of Li^- beams. Properties of particular concern were the photodetachment cross sections, energy levels, and autodetachment lifetimes. Detailed studies have been made of resonant structures in the photodetachment of He^- and Li^- negative ions. Some previously reported metastable ions were found to be probably nonexistent, while others were given the first experimental proof, and one unpredicted species (He_2^-) was discovered. A long-lived autodetaching form of OH^- was also observed. Total neutralization cross sections were measured as functions of beam energy for Li^+ in Mg, Ca, Sr, and Ba, and cross sections were calculated for Li^+ + Ca.

20. DISTRIBUTION/AVAILABILITY OF ABSTRACT UNCLASSIFIED/UNLIMITED ☒ SAME AS RPT. ☐ DTIC USERS ☐	21. ABSTRACT SECURITY CLASSIFICATION
22a. NAME OF RESPONSIBLE INDIVIDUAL Major Bruce L. Smith	22b. TELEPHONE NUMBER (Include Area Code) (202) 767-4908
22c. OFFICE SYMBOL NP	

DD FORM 1473, 83 APR EDITION OF 1 JAN 73 IS OBSOLETE.

Unclassified

SECURITY CLASSIFICATION OF THIS PAGE

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I RESEARCH OBJECTIVES

The main objective of this research was to determine the important properties of metastable autodetaching negative ions and means of producing them in beams. These ions are unique among negative ions because they are electronically excited above the ground state of the neutral parent, which itself cannot form a negative ion. They are metastable against autodetachment, however, because their spin configurations forbid E1 transitions to the ground state continuum. Depending on whether decay occurs through spin-orbit or spin-spin coupling and depending on the electronic complexity, the autodetachment lifetimes can vary from 10^{-7} to 10^{-4} s.

The use of these negative ions to form low-divergence high energy neutral beams is an interesting possibility because their self-neutralization by autodetachment (after acceleration) could be > 90% efficient without inducing any significant angular spread ($\sim 1 \times 10^{-6}$ radian). These species also have potentially even more attractive properties for photoneutralization because of the photodetachment resonances that are expected to occur near the higher excited state thresholds can be reached with existing lasers, which is not possible in the cases of most stable ions. Among these ions, only He^- was known when we began this work, but others (Be^-, N^-, Mg^-, and the molecules H_2^-, H_3^-, and N_2^-) had been either predicted theoretically or reported experimentally although their existence had never been fully confirmed.

The main barrier to studies of these ions in the past arose because the spin configurations that provide their metastability also inhibit their formation in conventional ion sources. However, their formation from a positive ion beam by two successive electron-capture collisions in an alkali metal vapor, which we implemented, is both allowed and effective for experimental studies. In addition to determining their properties (such as energy levels, lifetimes, and photodetachment cross sections), we also planned to investigate methods of improving the production of these ion beams by varying the alkali target and beam energy.

A second aspect of this research was both experimental and theoretical research on methods of producing Li^- from Li^+ beams. Li^- has been suggested as an attractive neutral beam parent for its collisional properties, but it also has a photodetachment "cusp," which had previously been calculated to reach 1.3 $Å^2$ at visible wavelengths.

The work has been very productive even though we have not yet completed all the tasks envisioned in the first proposal. As usually happens in basic research, we have encountered unexpected phenomena that led to additional important results and prompted both an expansion of the work and some digression from the original work plan.

II ACCOMPLISHMENTS

The accomplishments under this contract are summarized below. More complete descriptions of some of them can be obtained from past annual reports and from the publications cited.

1. First observation of the He^{-} $^4P^e$ Shape Resonance.[1] This difficult measurement required and achieved the first use of a cw dye laser at photon wavelengths in the region 900-1000 nm. It confirmed the predictions of Hazi and Reed,[2] which followed our preliminary survey of He^{-} photodetachment.[3] We located the resonance, measured the width, and found that the cross section reached the amazing size of ≈ 75 Å^2. Because of the weakness and instabilies of the laser at the longest wavelengths, we were unable to reach the threshold of the He 2^3P + e outgoing state; also, evidence of possible interference structures indicated that the measurements needed further improvement.

2. Construction and installation of electrostatic quadrupole deflectors to permit coaxial laser-ion beam experiments. This effort improved our signal/background ratio in photodetachment by about a factor of 50 and greatly improved subsequent measurements.

3. Calculation of Li^{+} + Ca charge transfer cross sections. In support of an early AFWL interest in Li^{-} production from Li^{+}, *ab initio* calculations were made by Olson and coworkers[4] under subcontract to SRI. These were eventually tested by measurements at SRI.

4. Measurements of Li^{+} charge transfer cross section in alkaline earth targets. These cross sections represent the first step in the production of Li^{-} from Li^{+} beams by two-step electron capture. Measurements were made for the first time in Mg, Ca, Sr, and Ba vapor targets at energies between 1 and 10 keV. A description of the results[5] has been published in *Physical Review A*.

5. Measurements of Li^- equilibrium yields in Mg, Ca, Sr, and Ba vapors. This work was performed at LBNL in a collaborative effort.[6] The results in Mg were suprising, but were made understandable by the results of our work on Li^+ charge transfer in Mg cited in 4 above.

6. Strong evidence was obtained against the existence of metastable H_2^- and H_3^-. This result, described in a paper in the Physical Review,[7] was disappointing but was nevertheless regarded as a positive contribution in resolving the conflict between the negative results of theory and the reported experimental observations. Null results obtained in our search at that time for N^-, N_2^-, and Mg^- are discussed further below in item 13.

7. Discovery of the unsuspected metastable ion He_2^-. This exciting discovery[8] came during our first attempts to produce Be^-. Although to our knowledge, He_2^- had not been previously contemplated and was thus a surprise, after considering the results of several tests, we were convinced of its existence and also understood how it was produced. The parent neutral is the metastable excimer $He_2\ {}^3\Sigma_u^+$, which (we now know) is easily formed by resonant charge transfer of He_2^+ in Cs. Our conjectures[8] about its electronic configuration and our measured lifetimes were subsequently confirmed in *ab initio* calculations by Michels[9] (also under AFOSR suppport).

8. Observation of the metastable Be^-. After learning that Be^+ beams can be easily generated by admitting CCl_4 into an ion source containing a little Be metal, it was easy to obtain a beam of Be^- using Cs in the charge transfer cell. We were then able[10] to prove its metastability, measure its decay rate at several energies to demonstrate the existence of more than one fine structure level, and briefly survey its photodetachment cross sections between 1.7 and 2.5 eV.

9. Resolution of the $He^-\ {}^4P^e$ shape resonance and the 2^3P channel threshold. The increased signal/background ratio resulting from the coaxial laser-ion beam arrangement shown in Figure 1, plus our improvements to the operation of the dye laser, made it possible to obtain data over the whole resonance with much less uncertainty than before and to extend the photon energy range downward to include the 2^3P threshold as well. The results are

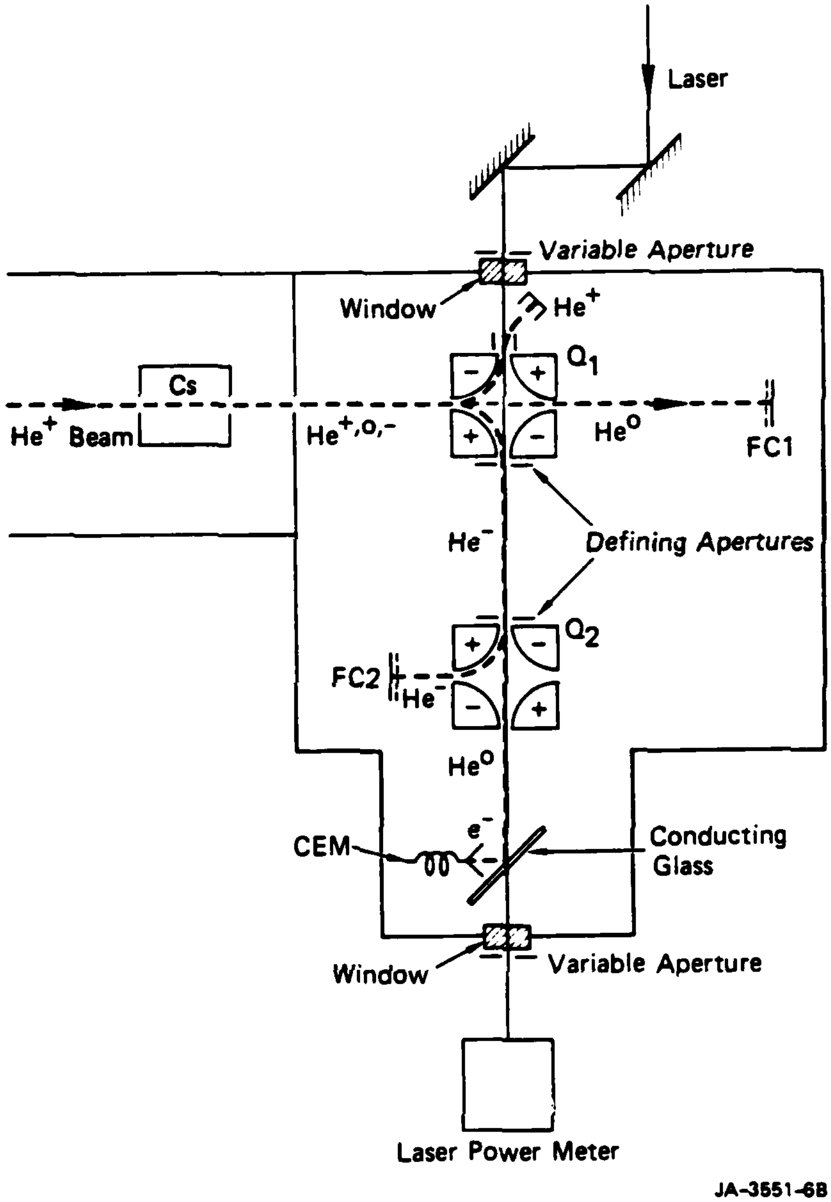

FIGURE 1

Coaxial laser-ion beam arrangement for measuring photodetachment cross sections and, with ion beam alone, autodetachment rates. The conducting glass plate permits detection of secondary electrons from neutral beam while simultaneously monitoring the transmitted laser power.

shown in Figure 2. The absolute values agree very well with our earlier crossed-beam measurements.

To our knowledge, this is the first electronic shape resonance that has been observed with such high resolution. As a result, our analysis of the threshold region, discussed below, showed deviations from the expected behavior and has led to new understanding of the effects of resonances on the behavior of cross sections near thresholds. These developments delayed for more than a year the completion of a paper describing the overall measurements and analysis of the resonance, but it is now in preparation for Physical Review.[11]

10. Analysis of the effects of a shape resonance on threshold behavior. An accurate determination of He 2^3P threshold energy E_0 in photodetachment can be used to determine the electron affinity of He 2^3S. However, we soon found that our cross sections did not fit the normal Wigner threshold law[12] $\sigma \propto k^{2\ell+1}$ (k and ℓ are the linear and angular momenta of the ejected electron, respectively), which becomes

$$\sigma \propto (E - E_0)^{\ell+1/2}$$

when $E = h\nu >> (E - E_0)$. The threshold energy E_0, given by least-squares fits of the data, continued to increase as we increased the number of data points included in the fit, even within 6 meV of the nominal E_0. Guessing that this formula is probably inadequate because it does not account for the proximity of the resonance, which causes a rapid increase in the cross section immediately above E_0, we sought a more comprehensive form. In fact, we took several approaches to the problem, but found a common solution. We found that not only the threshold region (Figure 3) but the entire resonance (Figure 3) could be fit quite well by the approximate parametric form

$$\sigma \propto \frac{(E - E_0)^{3/2}}{(E - E_r)^2 + (\Gamma/2)^2}$$

which is the simple product of the normal Wigner threshold law and the Breit-Wigner resonance form. However, the "width" Γ is a variable that, from the most explicit derivation, is proportional to $k^{2\ell+1} = (E - E_0)^{\ell+1/2}$.

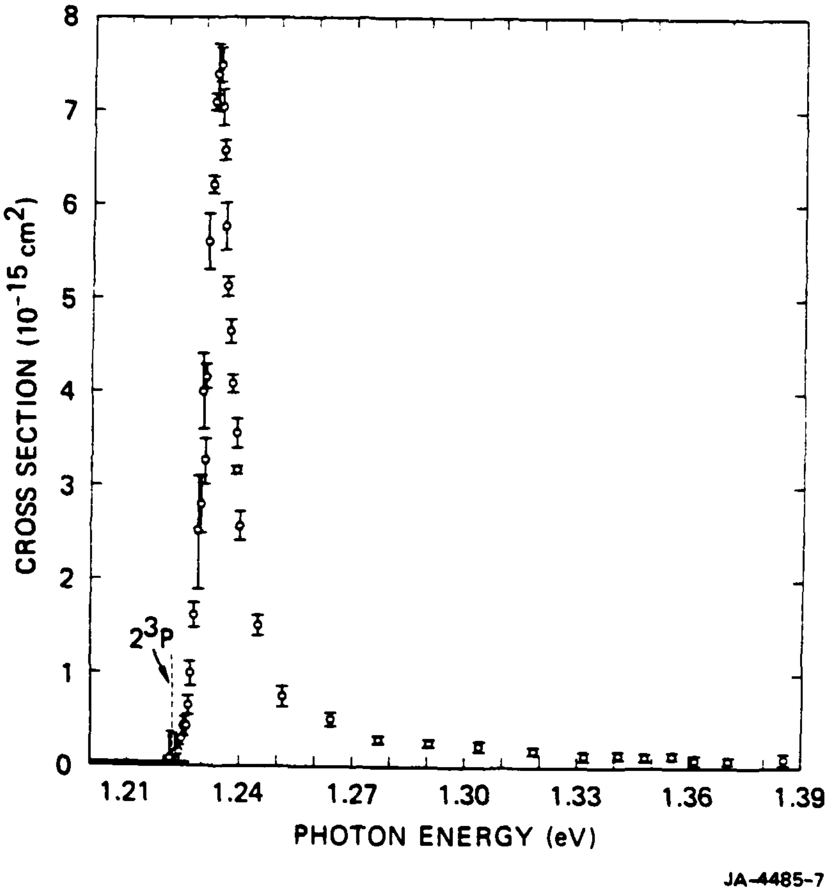

FIGURE 2

Total photodetachment cross sections for He^-, showing the $^4p^e$ shape resonance in high resolution.

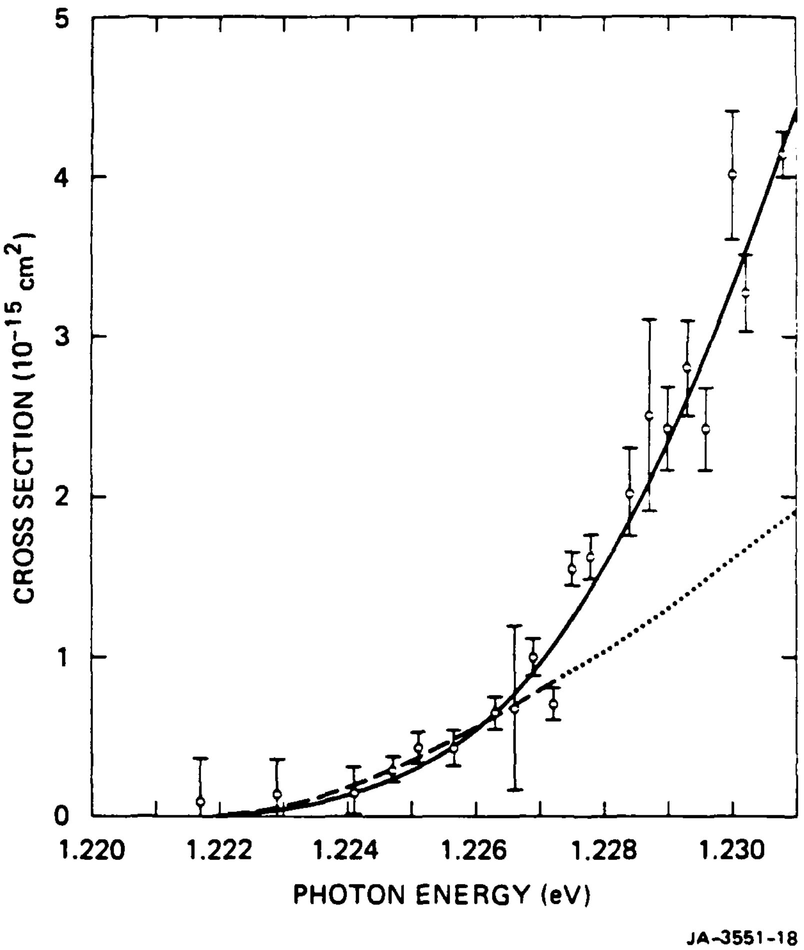

FIGURE 3

Photodetachment cross sections near the He(2^3P) + εp threshold. Dashed and dotted curve shows the Wigner threshold law fitted to the lowest 7 to 9 data points. Solid curve is the new modified threshold law fitted to all data shown. See text.

Fitting this form to all the data located the resonance E_r at 1234.3 ± 0.3 meV and the threshold E_o at 1221.9 ± 0.8 meV. The latter yields an electron affinity of 77.4 ± 0.8 meV for He 2^3S.

In spite of its simplicity, this is the first explicit form of a threshold law that accounts for the effects of a nearby resonance, and its success led to our analysis of the effects of other types of resonances on threshold behavior. A paper has now been accepted by Physical Review Letters.[13]

11. Measurement of the Li^- photodetachment cross section near the Li 2^2P threshold and analysis of the effects of a virtual resonance. Because of its intrinsic importance and its possible use in photodetachment neutralization of Li^- beams, we measured the Li^- photodetachment cross section in the region of the Li $2^2P + \varepsilon s$ threshold to examine the "Wigner cusp" that is theoretically expected to exist at the opening of any s-wave channel ($\ell = 0$). Again, we found that the normal formulae for describing a Wigner cusp was inadequate within a few meV above threshold. The effects of a resonance were also observed in the ab initio calculated cross sections of Moores and Norcross,[14] who found that the Wigner laws failed within 0.027 meV.

We found that the deviations are the effects of a "virtual state," which can occur in s-wave channels when the attractive correlation force is insufficient to form a "bound" or Feshbach resonance below the threshold. Using two-channel scattering theory, we were able to derive parametric forms to include these effects of the virtual resonance, both below and above the 2^2P threshold (see Figure 4). The data fitting yielded characteristic parameters of the resonance for comparison with theory. They included an accurate value of the threshold energy, which allowed us to obtain the Li electron affinity to an accuracy of 0.7 meV. A description of this work has been accepted by Physical Review as a Rapid Communication.[15]

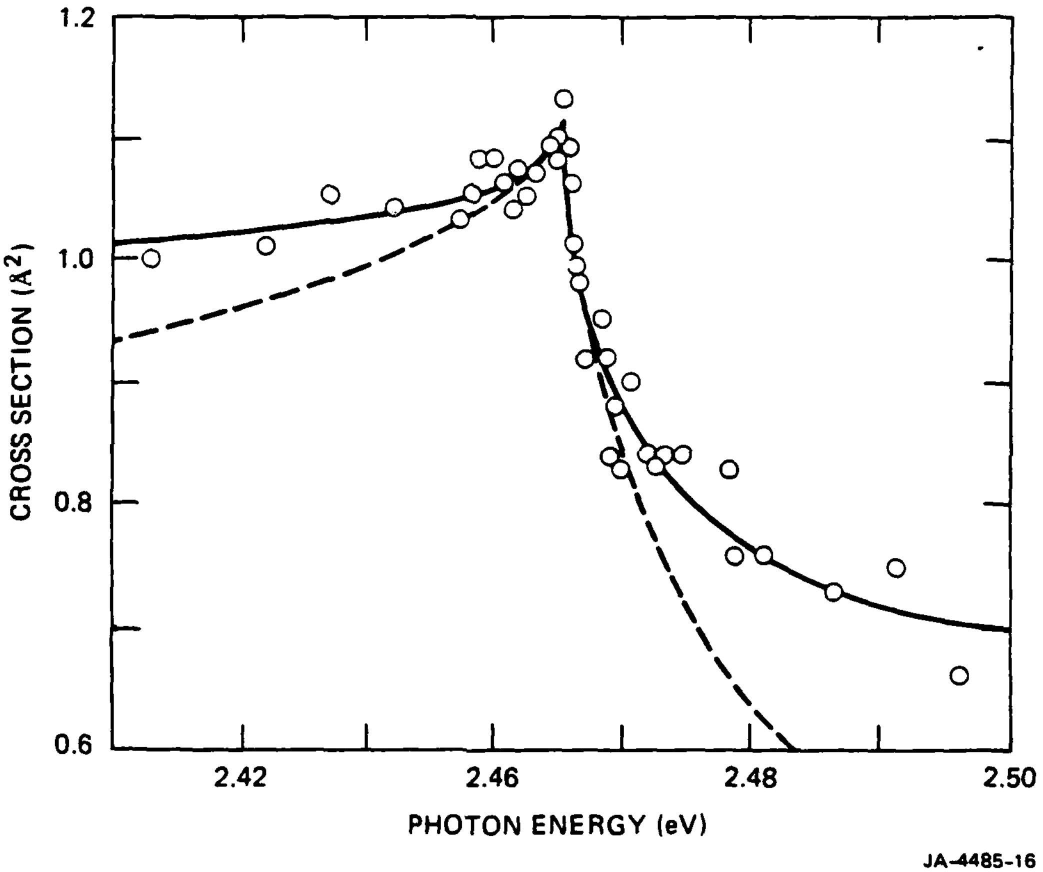

FIGURE 4

Li^- photodetachment cross section near the $Li(2^2P)$ threshold. Measured data are open circles. Solid curve shows our modified near-threshold formulae fitted to the data above and below the threshold. Dashed curve gives the best fit of the normal Wigner law to the measured data.

12. Analysis of the effects of a Feshbach resonance on opening-channel threshold behavior. Stimulated by the results described above, we considered the effects of the known[16] Feshbach resonances below the first excited ^2P-state thresholds in Rb and Cs. Rapid deviations of the ^2P channel partial cross sections had already been observed in photodetachment.[17,18] In Cs, these deviations occurred within 0.025 meV of the threshold.[18] Treating this problem with single-channel effective-range theory, we were able to derive a modified threshold law that fit these existing data very successfully for the first time.

These threshold analyses were all based on existing theory, but our application of it to explain the deviations from the Wigner law is new. The parametric formulae permit analyses of photodetachment and electron scattering data to yield information on the resonances that cause the deviations. Such analyses can now provide important details of the resonances for comparison with theory, thus increasing the value of the experimental data. An abstract[19] on "Modified Photodetachment Threshold Behavior Near Resonances" was submitted as a contributed paper to the XIV International Conference on the Physics of Electronic and Atomic Collisions (ICPEAC) in Palo Alto, California, July 1985. It was then upgraded to an Invited "Hot Topic" Paper for oral presentation, which will appear in the published book of Invited Papers.[20]

13. First observation of the metastable ion Ar^-, and measurement of its autodetachment lifetime. During our search last year for other metastable negative ions, we also looked for metastable Ne^- and Ar^- because analogous metastable core-excited autoionizing states exist in their isoelectronic relatives Na and K. We later learned that a metastable $3p^5\,4s4p\ ^4S$ state of Ar^- had recently been predicted by Bunge et al.[21] Because its production was probably inhibited by the energy limitations of our apparatus, we arranged to perform experiments on a higher energy apparatus at Lawrence Berkeley Laboratory (LBL).

We were successful in observing Ar^- produced from Ar^+ in Cs, and we measured its autodetachment rate at several beam energies (delay times after formation). These measurements indicated a single lifetime of 350 ± 150 ns and thus supported the ^4S state predicted by Bunge et al., which would have a

single J = 3/2 fine-structure state. A search for Ne^- indicated that no metastable states exist for that ion, also in agreement with the theoretical predictions of Bunge et al.[21] A manuscript on this work has been published in Physical Review Letters.[22]

We also conducted a second search for N^- and N_2^- at the higher beam energies available at LBL during these experiments, but again obtained null results that are fairly convincing that these ions do not exist, and that the original observations[23] were thus in error. We will submit a Brief Report on this work to the Physical Review. We had intended to search again for Mg^-, however the ion source available at LBL was not capable of producing the parent Mg^+ beam.

14. Measurement of autodetached and photodetached electron spectra. These measurements yield information on the energy levels of the negative ions and on the energies of the final neutral states. We chose to use the experimental arrangement shown in Figure 5 of its practical simplicity as well as its potentially high sensitivity. Not shown are the magnetic shields that surround (1) the energy analyzer and (2) the drift path between and the energy analyzer the electrostatic quadrupole Q_1. These mu-metal shields are required to reduce the ambient magnetic field from the earth (~ 0.3 Gauss) and other sources (~ 0.1 Gauss) to < 0.01 Gauss so that low-energy electron trajectories are undisturbed.

The apparatus in Figure 5 has been installed and tested, and we have encountered several difficulties in the process. Calibration measurements were made on He^- autodetached electrons and on photodetached electrons from H^-, using several photon energies from an argon ion laser. We have not yet reached a consistent understanding of the behavior of the system in the measurement of low-energy electrons. Such measurements are complicated by small contact potentials and stray electrical and magnetic fields, and our first measurements on the autodetached electrons from Be^- were not definitive enough to warrant publication.

However, we have also performed measurements on He_2^-, where the electron energies are higher and less subject to small field effects. The results show a broad peak, centered at about 15.6 eV, resulting from autodetachment to the repulsive He_2 $(^1\Sigma)$ ground-state potential. These results indicate that either the R_e of He^- calculated by Michels is too small by ~ 0.1 Å or the semiempirical curves of Foreman et al.[24] for the He_2 $(^1\Sigma)$ potential is too high by ~ 0.3 eV at R = 1.05 Å. This problem has not yet been resolved, and more effort will be given to it before the results are submitted for publication.

15. Observation of metastable autodetaching Ca^-. After several attempts, easily measurable currents (> 10 pA) of Ca^- were produced in our low-energy apparatus and were observed to autodetach. Apparent decay rates were measured, indicating lifetimes of about 1 ms. The slow decay rates have apparently prevented the detection of autodetachment in previous experiments in other laboratories. Planned upgrading of our bending magnet will permit more definitive experiments to be performed at higher energies in future work.

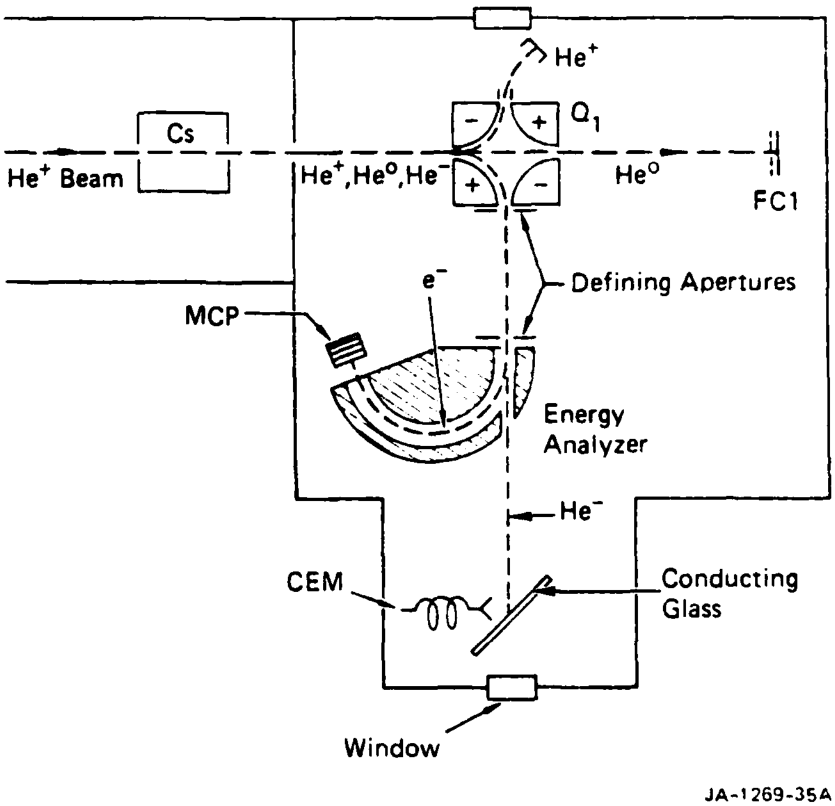

FIGURE 5

Apparatus arrangement for measuring the energy spectra of autodetached and photodetached electrons.

16. <u>Observation of inhibited autodetachment in OH^-</u>. In the search for Ca^- described above, we also found an autodetaching ion at mass 17, which was created when H_2O^+ ions (produced from a contaminant in the ion source) entered the alkali oven. The mass of the negative ion was deduced from the potentials on Q1 (Figure 1) required to deflect the autodetaching beam to the aperture in front of Q2. It was first conjectured that this ion was an electronically excited quintet state, and an abstract for a contributed paper was submitted to the XIV ICPEAC.[25] Later, energy measurements attempted on the autodetached electrons failed to show the expected peak at several eV. Also, consideration of the energetics of production from H_2O^+ by two-step electron capture indicated that an electronically excited state of OH^- was unlikely to be formed. We therefore suspected that vibrationally excited OH, produced by dissociative charge transfer in the first step, might generate OH^- in high v'' that are above $v' = 0$ of OH, and thus could autodetach. To test this hypothesis, we have used a technique we developed[26] to measure the kinetic energies released in dissociative charge transfer. We were able to conclude from those measurements that at least 2.5 eV internal energy is stored in the OH that forms the negative ions. Because states with $v'' > 5$ lie above the 1.83 eV electron affinity of OH and can autodetach, this seems to be the explanation. Rapid autodetachment is prevented because the OH and OH^- potentials are nearly identical except for their vertical displacement, and the off-diagonal Franck-Condon factors are very small. we have calculated these F-C factors and found that they are less than about 10^{-6} for autodetachment from $v'' > 5$. A more detailed analysis is being made in preparation for the publication of these results.

III PUBLICATIONS AND CONFERENCE PRESENTATIONS

The following paper and conference presentations were produced under this contract.

1. J. R. Peterson, M. J. Coggiola, and Y. K. Bae, "He^- Photodetachment: Effects of the 1s2p2p' $^4P^e$ Resonance," invited talk at the 7th Conference on the Applications of Accelerators in Research and Industry, North Texas State University, November 1982; IEEE Trans. Nucl. Sci. NS30 (2), 1043 (April 1983).

2. J. R. Peterson, M. J. Coggiola, and Y. K. Bae, "Measurement of the 1s2p2p'$^4P^e$ Resonance in the He^- Photodetachment," Phys. Rev. Lett. 50, 664 (1983).

3. J. R. Peterson, "Photoexcitation of Autodetaching Negative Ion Resonances," invited talk at the DEAP in Boulder, CO, May 1983; Bull. Am. Phys. Sci. 28, 779 (1973).

4. Y. K. Bae, M. J. Coggiola, and J. R. Peterson, "Search for D_2^-, D_3^-, and Other Metastable Ions Formed by Electron Capture in Alkali Vapors," presented at the Third International Symposium on the Production and Neutralization of Negative Ions and Beams," Brookhaven National Laboratory, November 14-18, 1983.

5. Y. K. Bae, M. J. Coggiola, and J. R. Peterson, "Observation of the Molecular Helium Negative Ion He_2^-," Phys. Rev. Lett. 52, 747 (1984).

6. Y. K. Bae, M. J. Coggiola, and J. R. Peterson, "A Search for H_2^-, H_3^-, and Other Metastable Negative Ions," Phys. Rev. A 29, 2888 (1984).

7. J. R. Peterson, Y. K. Bae, and D. L. Huestis, "Threshold Behavior Near an Electronic Shape Resonance; Analysis of the He(2^3P) Threshold in He^- Photodetachment and Determination of the He(2^3P) Electron Affinity," Phys. Rev. Lett. 55, 692 (1985).

8. J. R. Peterson, Y. K. Bae, and M. J. Coggiola, "Resolution of the He^- $^4P^e$ Shape Resonance and Total Photodetachment Cross Section and Analysis," in preparation for Phys. Rev. A.

9. M. J. Coggiola, Y. K. Bae, and J. R. Peterson, "Electron Capture by Li^+ in Mg, Ca, Sr, and Ba," Phys. Rev. A. 32, 784 (1985).

10. Y. K. Bae and J. R. Peterson, "Effects of a Virtual-State Resonance on Li^- Photodetachment Near the Li (2^3P) Threshold," accepted by Phys. Rev. Lett. A (1985).

11. Y. K. Bae and J. R. Peterson, "Observation of the Metastable Negative Beryllium Ion, Be^- ($^4P^e$)," Phys. Rev. 30, 2145 (1984).

12. J. R. Peterson, Y. K. Bae, M. J. Coggiola, and D. L. Huestis, "Details of the He^- $^4P^e$ Shape Resonance and Its Effect on Photodetachment Near the He 2^3P Threshold," presented at the 15th Annual Meeting of the Division of Electron and Atomic Physics in Storrs, Connecticut, May 30 - June 1 1984.

13. Y. K. Bae, M. J. Coggiola, and J. R. Peterson, "Observation of a Molecular Helium Negative Ion, He_2^-," presented at the 15th Annual Meeting of the Division of Electron and Atomic Physics in Storrs, Connecticut, May 30 - June 1 1984.

14. J. R. Peterson, Y. K. Bae, and D. L. Huestis, "Threshold Behavior Near An Electonic Shape Resonance," presented at the Ninth International Conference on Atomic Physics, Seattle, WA, July 21-27, 1984.

15. Y. K. Bae, J. R Peterson, A. S. Schlachter, and J. W. Stearns, "Observation of the Metastable Negative Argon Ion Ar^-," Phys. Rev. Lett. 54, 789 (1985); also presented at the DEAP, Norman OK (1985), Bull. Am. Phys. Soc. 30, 878 (1985).

16. J. R. Mowatt, E. Fisch, A. S. Schlachter, J. W. Stearns, and Y. K. Bae, "Equilibrium Charge State Fraction of Li^-, Li^o, and Li^+ in Mg, Sr, and Cs Vapors," Phys. Rev. A 31, 2893 (1985).

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19. Y. K. Bae and J. R. Peterson, in "Electronic and Atomic Collisions," abstracts of contributed papers, XIV Int. Conf. Phys. Elect. Atomic Coll., Palo Alto (1985), p. 48.

20. Y. K. Bae and J. R. Peterson, XIV ICPEAC Invited Papers, (North Holland, Amsterdam, 1986), to be published.

21. Y. K. Bae and J. R. Peterson, XIV ICPEAC, Abstracts of Contributed Papers, Palo Alto (July, 1984), p. 584.

22. Y. K. Bae, P. C. Cosby, and J. R. Peterson, "Observation of Inhibited Autodetachment in Excited OH^-," to be submitted to Chem. Phys. Lett.

IV PROFESSIONAL PERSONNEL

Y. K. Bae, M. J. Coggiola, and J. R. Peterson were primarily involved with this effort, but important contributions were also made by P. C. Cosby, H. Helm, and D. L. Huestis.

V INTERACTIONS

In addition to the contributed and invited conference presentations listed above, colloquia were also presented at Lawrence Livermore National Laboratory (LLNL) and Lawrence Berkeley Laboratory (LBL). Active collaboration with LLNL personnel has taken place in the successful search for Ar^{-22} and in determining the charge state fractions of Li^-, Li^0, and Li^+ in Mg, Sr, and Cs vapors.[6] M. J. Coggiola and J. R. Peterson both participated in the Whitehorse Ion Source Workshop at Los Alamos National Laboratory (LANL) in 1982; Dr. Peterson served as discussion leader for the "Atomic Physics of Ion Sources" session.

Consultations have also been held with Principal Investigators regarding AFOSR-supported work at the Jet Propulsion Laboratory and Hughes Aircraft. Active communication of our He_2^- discovery to H. Michels (United Technology Research) led to his calculation, also under AFOSR support, which was the first theoretical demonstration of its existence and study of its characteristics.

VI NEW DISCOVERIES

In performing this work, we made the following discoveries:

(1) Discovered the unexpected ion He_2^-.

(2) Found and defined the sources of the deviations from the Wigner threshold law that appeared in our work near the He^- shape resonance as well as those that had been observed in previous photodetachment experiments on Rb^- and Cs^- (near Feshbach resonances) and in ab-initio calculations on Li^-(near a virtual state).

(3) Found the metastable ion Ar^-, which had not been observed previously.

(4) Found that OH^- has long-lived autodetaching states.

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